

CLAIMS

1. A negative electrode for lithium secondary batteries, comprising a negative active material and a binder,

5 wherein the negative active material comprises graphite A and graphite B,

shapes of primary particles of the graphite A are spherical or elliptical,

an average particle diameter of the primary particles of the graphite

10 A ranges between 10  $\mu\text{m}$  and 30  $\mu\text{m}$  inclusive,

sizes of crystallites of the graphite A in a direction of a c-axis are smaller than 100 nm and tap density of the graphite A is 1.0 g/cm<sup>3</sup> or higher,

shapes of primary particles of the graphite B are flat,

an average particle diameter of the primary particles of the graphite

15 B ranges between 1  $\mu\text{m}$  and 10  $\mu\text{m}$  inclusive, and

sizes of crystallites of the graphite B in a direction of a c-axis are 100 nm or larger.

2. The negative electrode for lithium secondary batteries according to

20 Claim 1, wherein at least a part of surfaces of the graphite A is further covered with non-graphite carbon.

3. The negative electrode for lithium secondary batteries according to

Claim 1,

25 wherein,  $I_{1350}$  denotes Raman intensity at approximately 1350cm<sup>-1</sup>,  $I_{1580}$  denotes Raman intensity at approximately 1580cm<sup>-1</sup> and a R-value of Raman spectrum is obtained by a formula:  $R = (I_{1350}/I_{1580})$ ,

a R-value of Raman spectrum of the graphite A is 0.4 or larger when the graphite A is excited by an Ar laser with a wavelength of 5145 Å.

4. The negative electrode for lithium secondary batteries according to Claim 1, wherein the primary particles of the graphite B aggregate or bond so as to form secondary particles, and an average particle diameter of the secondary particles ranges between 10  $\mu\text{m}$  and 30  $\mu\text{m}$  inclusive.

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5. The negative electrode for lithium secondary batteries according to Claim 1, wherein a weight proportion of the graphite A ranges between 10 wt% and 90 wt% inclusive, with respect to a sum weight of the graphite A and the graphite B.

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6. The negative electrode for lithium secondary batteries according to Claim 1, wherein the binder comprises a mixture of an aqueous resin and a rubber-based resin.

15 7. A method for manufacturing a negative electrode for lithium secondary batteries comprising the steps of:

preparing graphite A of which shapes of primary particles are spherical or elliptical, an average particle diameter of the primary particles ranges between 10  $\mu\text{m}$  and 30  $\mu\text{m}$  inclusive, sizes of crystallites in a direction of a c-axis are smaller than 100 nm, and tap density is 1.0 g/cm<sup>3</sup> or higher;

20 preparing graphite B of which shapes of primary particles are flat, an average particle diameter of the primary particles ranges between 1  $\mu\text{m}$  and 10  $\mu\text{m}$  inclusive, and sizes of crystallites in a direction of a c-axis are 100 nm or larger;

25 preparing paint by mixing the graphite A and the graphite B in the presence of a binder and a solvent; and

applying the paint on a collector, drying the paint and then performing a pressure forming treatment.

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8. The method for manufacturing the negative electrode for lithium secondary batteries according to Claim 7, wherein at least a part of surfaces of the graphite A is further covered with non-graphite carbon.
- 5 9. The method for manufacturing the negative electrode for lithium secondary batteries according to Claim 7,  
wherein,  $I_{1350}$  denotes Raman intensity at approximately  $1350\text{cm}^{-1}$ ,  $I_{1580}$  denotes Raman intensity at approximately  $1580\text{cm}^{-1}$  and a R-value of Raman spectrum is obtained by a formula:  $R=(I_{1350}/I_{1580})$ ,
- 10 a R-value of Raman spectrum of the graphite A is 0.4 or larger when the graphite A is excited by an Ar laser with a wavelength of  $5145\text{\AA}$ .
10. The method for manufacturing the negative electrode for lithium secondary batteries according to Claim 7, wherein the primary particles of the graphite B aggregate or bond so as to form secondary particles, and an average particle diameter of the secondary particles ranges between  $10\text{ }\mu\text{m}$  and  $30\text{ }\mu\text{m}$  inclusive.
- 20 11. The method for manufacturing the negative electrode for lithium secondary batteries according to Claim 7, wherein a weight proportion of the graphite A ranges between 10 wt% and 90 wt% inclusive, with respect to a sum weight of the graphite A and the graphite B.
- 25 12. The method for manufacturing the negative electrode for lithium secondary batteries according to Claim 7, wherein the binder comprises a mixture of an aqueous resin and a rubber-based resin.
13. A lithium secondary battery, comprising a positive electrode, a negative electrode and nonaqueous electrolyte,
- 30 wherein the negative electrode comprises a negative active material

and a binder,

the negative active material comprises graphite A and graphite B,  
shapes of primary particles of the graphite A are spherical or  
elliptical,

5 an average particle diameter of the primary particles of the graphite  
A ranges between 10  $\mu\text{m}$  and 30  $\mu\text{m}$  inclusive,

sizes of crystallites of the graphite A in a direction of a c-axis are  
smaller than 100 nm and tap density of the graphite A is 1.0 g/cm<sup>3</sup> or higher,  
shapes of primary particles of the graphite B are flat,

10 an average particle diameter of the primary particles of the graphite  
B ranges between 1  $\mu\text{m}$  and 10  $\mu\text{m}$  inclusive, and

sizes of crystallites of the graphite B in a direction of a c-axis are 100  
nm or larger.

15 14. The lithium secondary battery according to Claim 13, wherein at  
least a part of surfaces of the graphite A is further covered with  
non-graphite carbon.

15. The lithium secondary battery according to Claim 13,  
20 wherein,  $I_{1350}$  denotes Raman intensity at approximately 1350cm<sup>-1</sup>,  
 $I_{1580}$  denotes Raman intensity at approximately 1580cm<sup>-1</sup> and a R-value of  
Raman spectrum is obtained by a formula:  $R=(I_{1350}/I_{1580})$ ,  
a R-value of Raman spectrum of the graphite A is 0.4 or larger when  
the graphite A is excited by an Ar laser with a wavelength of 5145 Å.

25 16. The lithium secondary battery according to Claim 13, wherein the  
primary particles of the graphite B aggregate or bond so as to form  
secondary particles, and an average particle diameter of the secondary  
particles ranges between 10  $\mu\text{m}$  and 30  $\mu\text{m}$  inclusive.

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17. The lithium secondary battery according to Claim 13, wherein a weight proportion of the graphite A ranges between 10 wt% and 90 wt% inclusive, with respect to a sum weight of the graphite A and the graphite B.

5 18. The lithium secondary battery according to Claim 13, wherein the binder comprises a mixture of an aqueous resin and a rubber-based resin.